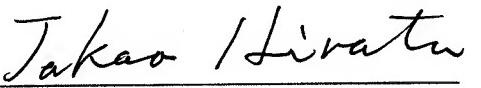


RE: U. S. APPLICATION NO. 10/507,320

I, Takao HIRATA of c/o TASHIRO & ETOH PATENT BUREAU of 8-18, Kyobashi 2-chome, Chuo-ku, Tokyo, Japan hereby declare that I am the translator of the concerned priority documents and certify that the followings is a true translation to the best of my knowledge and belief.

Signature of translator:



Takao HIRATA

Dated June 6, 2008

[Title of Document]

Specification

[Title of Invention]

5 Photo-Curable Adhesive Sheet and Process for the Preparation of Optical  
Information Recording Medium Obtained by Using the Same

[Scope of Claims for a Patent]

- [Claim 1] A photo-curable adhesive sheet comprising a photo-curable composition which comprises a reactive polymer having a photopolymerizable functional group and which has a glass transition temperature of not more than 20°C, the photo-curable adhesive sheet having a light transmittance of not less than 70% in a wavelength range of 380 to 420 nm.
- 10 [Claim 2] The photo-curable adhesive sheet as defined in claim 1, wherein the reactive polymer has a glass transition temperature of not more than 20°C.
- 15 [Claim 3] The photo-curable adhesive sheet as defined in claim 1 or 2, which has a light transmittance of not less than 80% in a wavelength range of 380 to 420nm.
- 20 [Claim 4] The photo-curable adhesive sheet as defined in any of claims 1 to 3, which has a light transmittance of not less than 70% in a wavelength range of 380 to 800nm.
- [Claim 5] The photo-curable adhesive sheet as defined in any of claims 1 to 4, wherein the reactive polymer is an acrylic resin.
- 25 [Claim 6] The photo-curable adhesive sheet as defined in any of claims 1 to 5, wherein the reactive polymer has 1 to 50% by mole of the

photopolymerizable functional group.

[Claim 7] The photo-curable adhesive sheet as defined in claim 6, wherein the photopolymerizable functional group is a (meth)acryloyl group.

5 [Claim 8] The photo-curable adhesive sheet as defined in any of claims 1 to 7, wherein the photo-curable composition contains 0.1 to 10% by weight of a photopolymerization initiator.

[Claim 9] The photo-curable adhesive sheet as defined in any of claims 1 to 8, which has a thickness of 5 to 300 $\mu\text{m}$ .

10 [Claim 10] The photo-curable adhesive sheet as defined in any of claims 1 to 9, wherein a release sheet is provided on at least one side of the photo-curable adhesive sheet.

[Claim 11] A process for the preparation of an optical information recording medium comprising:

15 superposing two optical information recording substrates having an uneven surface of recorded pits and/or grooves on each other through the photo-curable adhesive sheet as defined in any of claims 1 to 10 such that the two uneven surfaces face each other,

depressing the substrates and the adhesive sheet to form a laminate,

20 and

curing the laminate by light.

[Claim 12] The process as defined in claim 11, wherein the depressing step is carried out under reduced pressure.

25 [Claim 13] The process as defined in claim 11 or 12, wherein the depressing step is carried out at room temperature.

[Detailed Description of the Invention]

[Technical Field]

[0001]

The present invention relates to a photo-curable adhesive sheet advantageously utilized in a process for the preparation of an optical information recording medium, such as DVD (Digital Versatile Disc), an optical magnetic disc or a hard disc, in which a large amount of information such as letters, sound and animation is recorded and/or recordable as digital signals, and a process for the preparation of an optical information recording medium by using the sheet.

[Prior Art]

[0002]

As an optical information recording medium in which digital signals have been already recorded by forming pits on its surface, CD and CD-ROM are widely used. Recently, DVD that animation can be also recorded by forming pits on its both (double) sides has been noted as the next generation recording medium instead of CD and increasingly used. Further, attention is directed to recordable discs such as CD-R, DVD-R and DVD-RW having groove(s) or grooves and pits thereon. Furthermore, an optical magnetic disc and a hard disc are also well known as the recordable disc.

[0003]

The DVD readable from double sides can be prepared, for example, by subjecting melted polycarbonate resin to injection molding by the use of a stamper having unevenness (concave and convex) corresponding to the

reverse of unevenness of the signal-pit to be recorded on the substrate to prepare a transparent resin substrate having unevenness on its surface, forming a reflective layer on the uneven surface by spattering metal such as aluminum on it, and bonding two transparent resin substrates obtained in  
5 the above manner to each other through an adhesive such that the two reflective layers face each other. The adhesive usually uses an ultraviolet (UV) curable resin in the form of liquid.

[0004]

The preparation of the optical information recording medium such  
10 as DVD usually requires an adhesive step for bonding two transparent resin substrates as mentioned above. Since the optical information recording medium is recorded and read out by light, a substrate of the medium is needed to have a uniform thickness and high transparency, and be free from deformation such as warpage. When the liquid UV curable resin is used  
15 as an adhesive, the resultant adhesive layer has high transparency but is apt to have warpage due to large shrinkage on curing, which possibly results on reduction of dimensional stability.

[0005]

JA11-273147 describes that a pressure-sensitive adhesive sheet or a  
20 dry photopolymer in addition to the liquid UV curable resin is used for bonding a transparent film to an injection molded substrate having an uneven surface. However, the publication describes that the dry photopolymer is not preferred due to its low transparency.

[0006]

25 With increase of information to be recorded, a new optical information recording medium, which has larger storage capacity than DVD now on

use, is proposed. To obtain the large storage capacity, it is required to not only reduce sizes of signal pits and a groove but also shorten a wavelength of recording or reading laser. Further the shortening of the wavelength reduces distance between the laser and a surface having the pits and therefore it is needed to reduce a thickness of the optical recording substrate.

5 Hence, it is also preferred to reduce a thickness of the adhesive layer.

[Problem to be solved by the Invention]

[0007]

10 The present invention is made in view of the disadvantage of the above-mentioned adhesive, and hence the object of the invention is to provide a photo curable adhesive sheet that can be advantageously used in the preparation of an optical information recording medium.

[0008]

15 Further, the object of the invention is to provide a photo curable adhesive sheet having good dimensional stability and high transparency.

[0009]

20 Furthermore, the object of the invention is to provide a process for the preparation of an optical information recording medium by using the photo curable adhesive sheet.

[Means for Achieving Object]

[0010]

25 The object can be attained by a photo-curable adhesive sheet comprising a photo-curable composition which comprises a reactive polymer having a photopolymerizable functional group and which has a glass transi-

tion temperature of not more than 20°C, the photo-curable adhesive sheet having a light transmittance of not less than 70% in a wavelength range of 380 to 420 nm.

[0011]

5 In the photo-curable adhesive sheet, the reactive polymer preferably has a glass transition temperature of not more than 20°C, which renders the formation of unevenness by depression at room temperature easy. The photo-curable adhesive sheet preferably has a light transmittance of not less than 80% in a wavelength rang of 380 to 420nm. Further, the adhesive  
10 sheet preferably has a light transmittance of not less than 70% in a wavelength rang of 380 to 600nm, especially in a wavelength rang of 380 to 800nm. It is ensured that an optical disc (optical information recording medium) obtained by using the adhesive sheet having the light transmittance is read out without error if the disc is done by irradiation of laser.  
15 Moreover, the photo-curable adhesive sheet generally has cure shrinkage of not more than 8%.

[0012]

Further, it is preferred that the reactive polymer is an acrylic resin. Further the reactive polymer preferably has 1 to 50% by mole of the photopolymerizable functional group. The photopolymerizable functional group generally is a (meth)acryloyl group. The preferred reactive polymer is an acrylic resin having the photopolymerizable functional group through a urethane bond. The photo-curable composition generally contains 0.1 to 10% by weight of a photopolymerization initiator. The  
20 photo-curable adhesive sheet preferably has a thickness of 5 to 300 $\mu$ m. It is preferred that a release sheet is provided on at least (especially both  
25

sides) one side of the photo-curable adhesive sheet.

[0013]

The invention is also provided by a process for the preparation of an optical information recording medium comprising:

- 5       superposing two optical information recording substrates having an uneven surface of recorded pits and/or grooves on each other through the photo-curable adhesive sheet as defined above such that the two uneven surfaces face each other,  
            depressing the substrates and sheet to form a laminate, and  
10       curing the laminate by light.

[0014]

In the process, the depressing step is preferably carried out under reduced pressure, and the depressing step is also preferably carried out at room temperature.

- 15       [Best Mode for Conducting the Invention]

[0015]

Embodiments of the invention are explained in detail by referring to drawings.

- 20       [0016]  
            Fig. 1 is a section view showing an example of an embodiment of the photo-curable adhesive sheet according to the present invention.

[0017]

- 25       Fig. 1 is a section view showing an example of an embodiment of the photo-curable adhesive sheet 11 according to the present invention. In Fig. 1, the photo-curable adhesive sheet 11 has release sheets 12a, 12b on

its both sides. The release sheet may be provided only on one side of the photo-curable adhesive sheet, and otherwise may not be provided, depending on uses of the resultant optical disc. The provision on the both sides is advantageous because it facilitates the handling of the adhesive sheet.

5 [0018]

The photo-curable adhesive sheet 11 is a layer that is capable of deforming precisely along an uneven surface of, for example, an optical information substrate when the photo-curable adhesive sheet 11 is depressed on the uneven surface, and is mainly composed of a photo-curable composition which comprises a reactive polymer having a photopolymerizable functional group and which has a glass transition temperature of not more than 20°C. Further, the photo-curable adhesive sheet 11 has a light transmittance of not less than 70% in a wavelength range of 380 to 420 nm such that information can be easily read out by a reading (reproduction) laser. The light transmittance is preferably not less than 80% in a wavelength range of 380 to 420 nm. Hence, an optical disc obtained by using the adhesive sheet can be advantageously used in a process for reproducing pit signals by using a reproduction laser having a laser wavelength in the range of 380 to 420 nm.

20 [0019]

The photo-curable adhesive sheet of the invention has flexibility such that contact bonding can be carried out at room temperature as described above. Hence, the photo-curable adhesive sheet has excellent handling, and therefore can be widely used in not only information recording media such as CD, DVD, CD-R, DVD-R and DVD-RW but also the other various uses. The photo-curable adhesive sheet can be particu-

larly advantageously used in uses requiring precise bonding, such as the preparation of an electrical appliance, a furniture, an automobile, an instrument, a sports equipment or packing materials.

[0020]

5       The optical information recording medium can be prepared using the above photo-curable adhesive sheet, for example, as shown in Fig. 2.

[0021]

10      The release sheet 12a is removed from the photo-curable adhesive sheet 11 having release sheets 12a, 12b. The photo-curable adhesive sheet 11 having release sheet 12b is placed on an optical information recording substrate 21 having an uneven surface as recording pits and a reflective layer 23 (generally reflective layer of Al, Ag or the like having high reflectivity) provided on the uneven surface such that a side having no release sheet of the photo-curable adhesive sheet 11 faces the reflective layer, and 15 they are depressed. Thus, the side of the photo-curable adhesive sheet 11 closely adheres to the uneven surface of the reflective layer to form a laminate (consisting of 12b, 11, 23 and 21). The release sheet 12b is removed from the laminate.

[0022]

20      Subsequently, another optical information recording substrate 24 having an uneven surface as recording pits and a reflective layer 25 (or semitransparent reflective layer) provided on the uneven surface is placed on an uncured surface (having no substrate), where the release sheet 12b is removed, of the photo-curable adhesive sheet 11, such that the reflective layer 25 faces the uncured surface of optical information recording substrate 24, and they are depressed. Thus, the surface of the photo-curable 25

adhesive sheet 11 closely adheres to the uneven surface of the reflective layer of the substrate to form a laminate (consisting of 21, 23, 11, 24 and 25). The photo-curable adhesive sheet 11 of the laminate is cured by irradiation of ultraviolet rays. Thus the optical information recording medium is obtained.

[0023]

Otherwise, as shown in Fig. 3, a photo-curable adhesive sheet 11 having only a release sheet 12b is placed on an optical information recording substrate 24 having an uneven surface as recording pits and a reflective layer 23 provided on the uneven surface such that a side having no release sheet of the photo-curable adhesive sheet 11 faces the reflective layer. They are not depressed, and the release sheet 12b is removed from the adhesive sheet. Subsequently, another optical information recording substrate 24 is placed on a surface having no substrate of the photo-curable adhesive sheet 11, such that the reflective layer 25 faces the surface of the substrate 24, and then they are depressed at a time. Thus, the surface of the photo-curable adhesive sheet 11 closely adheres to the two uneven surfaces of the reflective layers of the substrates to form a laminate. The photo-curable adhesive sheet 11 of the laminate may be cured by irradiation of ultraviolet rays.

[0024]

In the above procedure, a polymer sheet for protection may be bonded onto the adhesive sheet instead of another optical information recording substrate 24. In this case, there is only one uneven surface in the laminate. Otherwise, a (photo-curable or adhesive) transfer sheet is placed on an optical information recording substrate having an uneven sur-

face as recording pits and a reflective layer provided on the uneven surface such that the transfer sheet faces the reflective layer. Subsequently, a stamper is depressed on the transfer sheet to form an uneven surface on the transfer sheet (if necessary cured), whereby a substrate having two uneven surfaces is prepared, and then a polymer sheet for protection may be bonded onto the exposed uneven surface through a photo-curable adhesive sheet of the invention.

[0025]

In the above process, the optical information recording medium exclusively used for reproduction is explained. However, an optical information recording medium used for recording (writing) is also prepared in the same manner as the above process. In the recordable medium, for example, grooves or grooves and pits is provided instead of the pits, and a metal recording layer is provided instead of the reflective or semitransparent reflective layer. When the recording layer is a dye-recording layer, a recording layer and reflective layer are generally provided. Besides these points, the recordable medium can be also prepared in the same manner as above.

[0026]

The optical information recording substrate may be prepared by a conventional injection molding or by the process comprising depressing a stamper on the photo-curable adhesive sheet of the invention, the adhesive sheet or the like. Thus, the optical information recording substrate of the invention can be prepared so as to have a thickness of 300 $\mu\text{m}$  or less.

[0027]

In the process, when the photo-curable adhesive sheet is depressed

on the optical information recording substrate, or when the two optical information recording substrate are superposed on each other through the photo-curable adhesive sheet such that the reflective layers face each other, it is preferred to carry out the depressing or superposing operation under 5 reduced pressure whereby bubbles generated in the operation can be easily removed.

[0028]

The depressing operation under the reduced pressure can be performed by a method comprising passing a substrate and photo-curable adhesive sheet or a substrate, photo-curable adhesive sheet and substrate, between two rolls under reduced pressure; or by a method comprising placing a substrate in a mold of a vacuum molding device and bring a photo-curable adhesive sheet into contact with the substrate under reduced pressure; or by a method comprising placing a substrate in a mold of a 10 vacuum molding device and bring a photo-curable adhesive sheet and substrate into contact with the substrate under reduced pressure. 15

[0029]

Further, the depressing operation under the reduced pressure can be performed using a device according to a double vacuum chamber system. 20 The operation is explained by referring to Fig. 4. Fig. 4 shows a laminator according to a double vacuum chamber system. The laminator is provided with a lower chamber 41, an upper chamber 42, a sheet of silicone rubber 43 and a heater 45. A laminate 49 consisting of a substrate having unevenness and a photo-curable adhesive sheet provided thereon (or laminate of substrate/adhesive sheet/substrate) is placed in the lower chamber 25 41 of the laminator. Both the upper chamber 42 and lower chamber 41

are degassed or decompressed. The laminate 49 is heated with a heater 45, and air is introduced into the upper chamber 42 to allow the chamber to be at atmospheric pressure while the lower chamber 41 is kept under reduced pressure, whereby the laminate is depressed to be contact bonded. After 5 cooling, the laminate is taken out and transformed to the next step. This operation permits sufficient deaeration under reduced pressure, and therefore, the substrate and the photo-curable adhesive sheet can be contact bonded without bubbles.

[0030]

10 The photo-curable adhesive sheet according to the invention comprises a photo-curable composition which comprises a reactive polymer having a photopolymerizable functional group and which has a glass transition temperature of not more than 20°C.

15 The photo-curable composition is generally composed mainly of the reactive polymer having a photopolymerizable functional group, a compound (e.g., monomer or oligomer) having a photopolymerizable functional group (preferably (meth)acryloyl group), a photopolymerization initiator and if necessary other additives.

[0031]

20 Examples of the reactive polymer having a photopolymerizable functional group include homopolymers or copolymers (i.e., acrylic resins having a photopolymerizable functional group) derived from alkyl acrylate (e.g., methyl acrylate, ethyl acrylate, butyl acrylate, 2-ethylhexyl acrylate) and/or alkyl methacrylate (e.g., methyl methacrylate, ethyl methacrylate, 25 butyl methacrylate, 2-ethylhexyl methacrylate) and having a photopolymerizable functional group on its main chain or side chain. These

(co)polymers can be obtained, for example, by copolymerizing one or more (meth)acrylate mentioned above with (meth)acrylate (e.g., 2-hydroxyethyl (meth)acrylate) having a functional group such as -OH and reacting the resultant polymer with a compound (e.g., isocyanatoalkyl (meth)acrylate)

- 5 having a functional group capable of reacting with the functional group of the polymer and having a photopolymerizable functional group. Thus an acrylic resin having a photopolymerizable functional group through a urethane bond is preferred.

[0032]

- 10 The reactive polymer of the invention has generally 1 to 50% by mole, preferably 5 to 30% by mole of the photopolymerizable functional group. Examples of the photopolymerizable functional group preferably include acryloyl, methacryloyl and vinyl groups, especially acryloyl and methacryloyl groups.

15 [0033]

- In case the reactive polymer having glass transition temperature of not more than 20°C is used as above, the resultant photo-curable transfer layer having flexibility can follow exactly the uneven surface of the stamper when the sheet is depressed on the stamper. The reactive polymer especially has glass transition temperature of 15 to -50°C because the resultant photo-curable layer can follow more exactly the uneven surface. When the glass transition temperature exceeds the upper limit, high pressure is needed in the depressing and bonding steps of the sheet, which brings about lowering of workability. When the glass transition temperature falls to below the lower limit, the resultant cured sheet does not have sufficient hardness.

## [0034]

The reactive polymer of the invention generally has number-average molecular weight of 5,000 to 1,000,000, preferably 10,000 to 300,000, and/or generally has weight-average molecular weight of 5,000 to  
5 1,000,000, preferably 10,000 to 300,000.

## [0035]

Examples of the compounds having a photopolymerizable group include (meth)acrylate monomers such as 2-hydroxyethyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate, 4-hydroxybutyl (meth)acrylate,  
10 2-ethylhexylpolyethoxy (meth)acrylate, benzyl (meth)acrylate, isobornyl (meth)acrylate, phenoxyethyl (meth)acrylate, tricyclodecane mono(meth)acrylate, dicyclopentenyloxyethyl (meth)acrylate, tetrahydrofurfuryl (meth)acrylate, acryloylmorpholine, N-vinylcaprolactam, 2-hydroxy-3-phenoxypropyl (meth)acrylate, o-phenylphenoxyethyl  
15 (meth)acrylate, neopentylglycol di(meth)acrylate, neopentyl glycol dipropoxy di(meth)acrylate, neopentyl glycol hydroxypivalate di(meth)acrylate, tricyclodecanedimethylol di(meth)acrylate, 1,6-hexanediol di(meth)acrylate, nonanediol di(meth)acrylate, trimethylolpropane tri(meth)acrylate, pentaerythritol tri(meth)acrylate, pentaerythritol tetra(meth)acrylate,  
20 tris[(meth)acryloxyethyl]isocyanurate and ditrimethylolpropane tetra(meth)acrylate; and

the following (meth)acrylate oligomer such as:

polyurethane (meth)acrylate such as compounds obtained by reaction of:

25 a polyol compound (e.g., polyol such as ethylene glycol, propylene glycol, neopentyl glycol, 1,6-hexanediol, 3-methyl-1,5-pentanediol,

1,9-nonanediol, 2-ethyl-2-butyl-1,3-propanediol, trimethylolpropane, diethylene glycol, dipropylene glycol, polypropylene glycol, 1,4-dimethylolcyclohexane, bisphenol-A polyethoxydiol and polytetramethylene glycol; polyesterpolyol obtained by reaction of the above-mentioned polyol and polybasic acid or anhydride thereof such as succinic acid, maleic acid, itaconic acid, adipic acid, hydrogenated dimer acid, phthalic acid, isophthalic acid and terephthalic acid; polycaprolactone polyol obtained by reaction of the above-mentioned polyol and  $\epsilon$ -caprolactone; a compound obtained by reaction of the above-mentioned polyol and a reaction product of the above-mentioned polybasic acid or anhydride thereof and  $\epsilon$ -caprolactone; polycarbonate polyol; or polymer polyol), and

an organic polyisocyanate compound (e.g., tolylene diisocyanate, isophorone diisocyanate, xylylene diisocyanate, diphenylmethane-4,4'-diisocyanate, dicyclopentanyl diisocyanate, hexamethylene diisocyanate, 2,4,4'-trimethylhexamethylene diisocyanate, 2,2',4'-trimethylhexamethylene diisocyanate), and

hydroxyl-containing (meth)acrylate (e.g., 2-hydroxyethyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate, 4-hydroxybutyl (meth)acrylate, 2-hydroxy-3-phenyloxypropyl (meth)acrylate, cyclohexane-1,4-dimethylolmono(meth)acrylate, pentaerythritol tri(meth)acrylate or glycerol di(meth)acrylate);

bisphenol-type epoxy(meth)acrylate obtained by reaction of bisphenol-A epoxy resin or bisphenol-F epoxy resin and (meth)acrylic acid.

These compounds having photopolymerizable functional group can be employed singly or in combination of two or more kinds.

[0036]

Any photopolymerization initiators known can be used in the invention. The initiators having good storage-stability after mixing with other components are preferred. Examples of the photopolymerization

5 initiators include acetophenone type initiators such as 2-hidroxy-2-methyl-1-phenylpropane-1-on,

1-hydroxycyclohexylphenylketone and

2-methyl-1-[4-(methylthio)phenyl]-2-morphorino-propane-1-on; benzoin type initiators such as benzylmethylketal; benzophenone type initiators

10 such as benzophenone, 4-phenylbenzophenone and hydroxybenzophenone;

thioxanthone type initiators such as isopropylthioxanthone and 2,4-diethythioxanthone. Further, as special type, there can be mentioned methylphenylglyoxylate. Especially preferred are

2-hidroxy-2-methyl-1-phenylpropane-1-on,

15 1-hydroxycyclohexylphenylketone,

2-methyl-1-[4-(methylthio)phenyl]-2-morphorinopropane-1-on and benzophenone. These photopolymerization initiators can be employed together

with one or more kinds of a photopolymerization promoter such as a benzoic acid type compound (e.g., 4-dimethylaminobenzoic acid) or a tertiary

20 amine compound by mixing the initiator with the promoter in optional ratio.

Only the initiator can be employed singly or in combination of two or more kinds. The initiator is preferably contained in the photo-curable composition in the range of 0.1 to 20% by weight, particularly 1 to 10% by weight.

[0037]

25 In addition to the above-mentioned photopolymerizable initiators, the acetophenone type initiator includes 4-phenoxydichloroacetophenone,

- 4-t-butyldichloroacetophenone, 4-t-butyltrichloroacetophenone,  
diethoxyacetophenone, 2-hidroxy-2-methyl-1-phenylpropane-1-on,  
1-(4-isopropylphenyl)-2-hidroxy-2-methylpropane-1-on,  
1-(4-dodecylphenyl)-2-hydroxy-2-methylpropane-1-on,  
5 4-(2-hydroxyethoxy)-phenyl(2-hydroxy-2-propyl)ketone,  
1-hydroxycyclohexylphenylketone,  
2-methyl-1-[4-(methylthio)phenyl]-2-morphorino-propane-1-on; and the  
benzophenone type initiators include benzophenone, benzoylbenzoic acid,  
methyl benzoylbenzoate, 4-phenylbenzophenone, hydroxybenzophenone,  
10 4-benzoyl-4'-methylphenylsulfide and  
3,3'-dimethyl-4-methoxybenzophenone.

## [0038]

The acetophenone type initiators preferably are  
2-hidroxy-2-methyl-1-phenylpropane-1-on,  
15 1-hydroxycyclohexylphenylketone,  
2-methyl-1-[4-(methylthio)phenyl]-2-morphorinopropane-1-on, and the  
benzophenone type initiators preferably are benzophenone, benzoylbenzoic  
acid and methyl benzoylbenzoate. Preferred examples of the tertiary  
amine compounds of the photopolymerization promoter include trietha-  
20 nolamine, methyldiethanolamine, triisopropanolamine,  
4,4'-dimethylaminobenzophenone, 4,4'-diethylaminobenzophenone, ethyl  
2-dimethylaminobenzonate, ethyl 4-dimethylaminobenzonate,  
(n-butoxy)ethyl 4-dimethylaminobenzonate, isoamyl  
4-dimethylaminobenzonate and 2-ethylhexyl 4-dimethylaminobenzonate.  
25 Especially preferred are ethyl 4-dimethylaminobenzonate, (n-butoxy)ethyl  
4-dimethylaminobenzonate, isoamyl 4-dimethylaminobenzonate and

2-ethylhexyl 4-dimethylaminobenzoate. As mentioned above, three kinds of components of the photopolymerizable initiators can be combined.

[0039]

The photo-curable composition of the invention is preferably configured such that the photo-curable transfer sheet has a glass transition temperature of not more than 20°C and the photo-curable transfer sheet has a transmittance of not less than 70% in a wavelength range of 380 to 420 nm. Therefore the photo-curable composition preferably contains, in addition to the compound having a photopolymerizable functional group and the photopolymerization initiator, if desired the following thermoplastic resin and other additives.

[0040]

The ratio by weight of the reactive polymer : the compound having a photopolymerizable functional group : the photopolymerization initiator generally is 40-100 : 0-60 : 0.1-10, especially 60-100 : 0-40 : 1-10.

[0041]

As other additives, a silane coupling agent can be used for enhancing the adhesive strength. Examples of the silane coupling agent include vinyltriethoxysilane, vinyltris(β-methoxyethoxy)silane, 20 γ-methacryloxypropylmethoxysilane, vinyltriacetoxysilane, γ-glycidoxypolytrimethoxysilane, γ-glycidoxypolytriethoxysilane, β-(3,4-epoxycyclohexyl)ethyltrimethoxysilane, vinyltrichlorosilane, γ-chloropropylmethoxysilane, γ-mercaptopropylmethoxysilane, γ-aminopropyltriethoxysilane, 25 N-β-(aminoethyl)-γ-aminopropyltrimethoxysilane. The silane coupling agent can be used singly, or in combination of two or more kinds. The si-

lane coupling agent is preferably used in the range of 0.01 to 5 weight by part based on 100 parts by weight of the above reactive polymer.

[0042]

Similarly, an epoxy group-containing compound can be used for enhancing the adhesive strength. Examples of the epoxy group-containing compounds include triglycidyl tris(2-hydroxyethyl)isocyanurate, neopentylglycol diglycidyl ether, 1,6-hexanediol diglycidyl ether, allyl glycidyl ether, 2-ethylhexyl glycidyl ether, phenyl glycidyl ether, phenol glycidyl ether, p-tert-butylphenyl glycidyl ether, diglycidyl adipate, diglycidyl o-phthalate, glycidyl methacrylate and butyl glycidyl ether. Further, the similar effect is also obtained by using an oligomer having an epoxy group and molecular weight of hundreds to thousands, or a polymer having an epoxy group and molecular weight of thousands to hundreds of thousands. The content of the compound having an epoxy group is sufficient in the range of 0.1 to 20 parts by weight based on 100 parts by weight of the reactive polymer, particularly 1 to 10% by weight. At least one of the compounds having an epoxy group can be used singly or in combination of two or more kinds.

[0043]

As other additives, further a hydrocarbon resin can be used for improving processing properties such as laminating properties. The hydrocarbon resin may be either natural resin or synthetic resin. Examples of the natural resins preferably include rosins, rosin derivatives and terpene resins. Examples of the rosins include gum resins, tall oil resins, wood resins. Examples of the rosin derivatives include hydrogenated rosins, disproportionated rosins, polymerized rosins, esterified rosins, metal

salts of rosins. Examples of the terpene resins include  $\alpha$ -pinene resins,  $\beta$ -pinene resins, and terpene phenol resins. Moreover, as the natural resin, dammar, copal, shellac can be used. Examples of the synthetic resins preferably include petroleum resins, phenol resins, and xylene resins.

- 5 Examples of the petroleum resins include aliphatic petroleum resins, aromatic petroleum resins, cycoaliphatic petroleum resins, copolymer type petroleum resins, hydrogenated petroleum resins, pure monomer type petroleum resins, and coumarone-indene resins. Examples of the phenol resins include alkylphenol resins and modified phenol resins. Examples of the  
10 xylene resins include xylene resins and modified xylene resins.

[0044]

Furthermore, acrylic resin can be employed in the invention. For example, homopolymers and copolymers obtained from alkyl acrylate(s) such as methyl acrylate, ethyl acrylate and butyl acrylate and/or alkyl  
15 methacrylate(s) such as methyl methacrylate, ethyl methacrylate and butyl methacrylate can be used. Copolymers of these monomers and other copolymerizable monomers can be also used. In view of reactivity in the photo curing step and durability and transparency of cured product, polymethyl methacrylate (PMMA) is preferred.

20 [0045]

The above-mentioned polymer such as hydrocarbon resin can be used in the amount of 1 to 20 parts by weight, preferably 5 to 15 parts by weight based on 100 parts by weight of the reactive polymer.

[0046]

25 The photo-curable composition may contain, in addition to the above-mentioned additives, an ultraviolet absorber, an aging resistant agent,

a dye, and a processing auxiliary agent in a small amount. If desired, particles of silica gel, calcium carbonate or silicone copolymer may be contained in a small amount.

[0047]

5       The photo-curable adhesive sheet comprising the photo-curable composition of the invention is generally prepared by homogeneously mixing the reactive polymer, a compound having a photopolymerizable functional group (monomer and oligomer) and if desired other additives, kneading the mixture using an extruder or roll, and subjecting the kneaded  
10 mixture to a film-forming process using a calendar, roll, T-die extrusion or inflation to form a film of a predetermined dimension. When a support is used, it is needed to form a film on the support. A more preferred process for forming the photo-curable sheet comprises the steps of: dissolving homogeneously the components in a good solvent, applying the resultant solution onto a separator coated closely with silicone or fluoric resin (or the support) by means of flow-coater method, roll-coater method, gravure-roll method, mayer-bar method or lip-die coating method, and vaporizing the solvent.  
15

[0048]

20       The surface of the photo-curable sheet may be embossed in the film formation process to prevent blocking and facilitate deaeration in the step depressing the sheet and the substrate or stamper. As methods for the embossing processing, conventional methods such as a method using embossing roll can be adopted. In a process for applying a solution, it is possible that the solution is applied onto an embossed film or paper having release properties whose emboss shape is transferred to the sheet. Mean  
25

surface roughness ( $R_a$ ) of the embossed surface is generally not more than 50 $\mu\text{m}$ , preferably 0.01 to 50 $\mu\text{m}$ , especially 0.1 to 20 $\mu\text{m}$ , whereby air is easily escaped from an interface between the sheet and a device to permit the embossed surface of the sheet to fill up complicated unevenness of the device. The mean surface roughness of less than 0.01 $\mu\text{m}$  is apt to bring about poor deaeration, whereas the mean surface roughness of more than 50 $\mu\text{m}$  occasionally allows the unevenness of the sheet to remain in the depressing step.

[0049]

The thickness of the photo-curable adhesive layer generally is in the range of 1 to 1,200 $\mu\text{m}$ , preferably 5 to 500 $\mu\text{m}$ , especially 5 to 300 $\mu\text{m}$ . When the thickness is thinner than 1 $\mu\text{m}$ , sealing properties are lowered and maybe the sheet does not full up the unevenness of the transparent substrate. When the thickness is thicker than 1,200 $\mu\text{m}$ , the thickness of the resultant recording medium is so thick whereby trouble in housing or storing of the medium and the resultant assembly or reverse influence in light transmittance possibly occurs.

[0050]

The release sheet used in the invention preferably comprises transparent organic resin having a glass transition temperature of not less than 50°C. The release sheet generally is a transparent resin sheet mainly consisting of organic resin such as polyester resin (e.g., polyethylene terephthalate, polycyclohexylene terephthalate, polyethylene naphthalate), polyamide (e.g., nylon 46, modified nylon 6T, nylon MXD6, polyphthalamide), ketone resin (e.g., polyphenylene sulfide, polythioether sulfone), sulfone resin (e.g., polysulfone, polyether sulfone), polyether nitrile, pol-

yarylate, polyether imide, polyamideimide, polycarbonate, polymethyl methacrylate, triacetylcellulose, polystyrene or polyvinyl chloride. Of these resins, polycarbonate, polymethyl methacrylate, polyvinyl chloride, polystyrene and polyethylene terephthalate can be preferably employed.

- 5 The thickness is generally in the range of 10 to 200 $\mu\text{m}$ , especially in the range of 30 to 100 $\mu\text{m}$ .

[0051]

The substrate having uneven surface used in the invention preferably comprises transparent organic resin having a glass transition temperature of not less than 50°C. The substrate generally is a transparent resin sheet mainly consisting of organic resin such as polyester resin (e.g., polyethylene terephthalate, polycyclohexylene terephthalate, polyethylene naphthalate), polyamide (e.g., nylon 46, modified nylon 6T, nylon MXD6, polyphthalamide), ketone resin (e.g., polyphenylene sulfide, polythioether sulfone), sulfone resin (e.g., polysulfone, polyether sulfone), polyether nitrile, polyarylate, polyether imide, polyamideimide, polycarbonate, polymethyl methacrylate, triacetylcellulose, polystyrene or polyvinyl chloride. Of these resins, polycarbonate, polymethyl methacrylate, polyvinyl chloride, polystyrene and polyethylene terephthalate are excellent in transferring properties and birefringence, and therefore can be preferably employed. The thickness is generally in the range of 200 to 2,000 $\mu\text{m}$ , especially in the range of 50 to 1,500 $\mu\text{m}$ .

[0052]

The material of the organic polymer film for protection preferably comprises transparent organic resin having a glass transition temperature of not less than 50°C. The film generally is a transparent resin sheet mainly

consisting of organic resin such as polyester resin (e.g., polyethylene terephthalate, polycyclohexylene terephthalate, polyethylene naphthalate), polyamide (e.g., nylon 46, modified nylon 6T, nylon MXD6, polyphthalamide), ketone resin (e.g., polyphenylene sulfide, polythioether sulfone),  
5 sulfone resin (e.g., polysulfone, polyether sulfone), polyether nitrile, polyarylate, polyether imide, polyamideimide, polycarbonate, polymethyl methacrylate, triacetylcellulose, polystyrene or polyvinyl chloride. Of these resins, polycarbonate, polymethyl methacrylate, polyvinyl chloride, polystyrene and polyethylene terephthalate are excellent in transparency  
10 and birefringence, and therefore can be preferably employed. The thickness is generally in the range of 10 to 200 $\mu\text{m}$ , especially in the range of 50 to 100 $\mu\text{m}$ .

[0053]

The photo-curable adhesive sheet of the invention obtained as  
15 above generally comprises the photo-curable composition containing the reactive polymer of a glass transition temperature of not more than 20°C. Further, the photo-curable adhesive layer generally has a light transmittance of not less than 70% in a wavelength rang of 380 to 800nm. In more detail, by setting the glass transition temperature of the reactive  
20 polymer to not more than 20°C, the resultant photo-curable adhesive sheet having flexibility can follow exactly the uneven surface of the substrate even at room temperature when the sheet is depressed on the stamper. Especially, in the case of the glass transition temperature of 15 to -50°C, the properties following exactly the uneven surface of the stamper is further  
25 improved. When the glass transition temperature is so high, high pressure and temperature is needed in the depressing or bonding operation whereby

the workability is reduced. When the glass transition temperature is so low, the resultant sheet after curing does not have sufficient hardness.

[0054]

As described above, the photo-curable adhesive sheet generally has  
5 a light transmittance of not less than 70% in a wavelength rang of 380 to 420nm, preferably 380 to 800nm, whereby reduction of the strength of signals to be read out with a laser beam can be prevented. Further, the sheet preferably has a light transmittance of not less than 80% in a wavelength rang of 380 to 420nm.

10 [0055]

The reactive polymer of the photo-curable composition preferably has 1 to 50% by mole of polymerizable functional group, whereby the cured photo-curable adhesive sheet has strength capable of holding its shape. The photopolymerization initiator is preferably used in the amount  
15 of 0.1 to 10% by weight as described previously. The amount of less than the lower limit causes workability to reduce owing to slow curing rate, whereas the amount of more than the upper limit causes the transfer precision to reduce.

[0056]

20 The photo-curable adhesive sheet of the invention can be offered as a film precisely controlled in the thickness, and therefore it is possible to easily and precisely bond the sheet to the uneven surface such as the substrate or stamper. This bonding can be easily carried out by depressing the sheet and stamper by means of easy method using pressure rollers or  
25 easy press to temporarily bond them at temperature of 20 to 100°C, and then curing the sheet by exposing it to light at room temperature for one to

tens seconds. Further, the temporarily bonded laminate is free from occurrence of slippage or peeling between of the sheet and stamper or substrate owing to its specific adhesion, and hence the laminate can be freely handled until the light-curing step.

5 [0057]

In case the photo-curable adhesive sheet of the invention is cured, it is possible to adopt, as light source used, various sources generating light in the wavelength range of ultraviolet to visible rays. Examples of the sources include super-high-pressure, high-pressure and low-pressure mercury lamps, a chemical lamp, a xenon lamp, a halogen lamp, a mercury halogen lamp, a carbon arc lamp, and an incandescent electric lamp, and laser beam. The exposing time is generally in the range of a few seconds to a few minutes, depending upon kinds of the lamp and strength of light.

[0058]

15 To promote the curing, the laminate may be heated beforehand for 30 to 80°C, and then the heated laminate may be exposed to ultraviolet rays.

[0059]

20 In the preparation of the optical information recording medium of the invention, it is continuously processed in the form of sheet and finally punched out in the form of disc. However, it may be processed in the form of disc when processing under reduced pressure is required.

[0060]

[EXAMPLE]

25 The invention is illustrated in detail using the following Examples.

[Example 1]

<Preparation of photo-curable adhesive sheet>

(Preparation of reactive polymer)

Formulation I

2-ethylhexyl methacrylate	70 parts by weight
5 methyl methacrylate	20 parts by weight
2-hydroxyethyl methacrylate	10 parts by weight
benzophenone	5 parts by weight
toluene	30 parts by weight
ethyl acetate	30 parts by weight

10 A mixture of the above Formulation I was heated to 60°C with moderately stirring to initiate the polymerization, and stirred at this temperature for 10 hours to provide acrylic resin having a hydroxyl group on its side chain. Then, 5 parts by weight of Calens MOI (2-isocyanatoethyl methacrylate; available from Showa Denko K.K.) was added to the solution 15 of the acrylic resin, and reacted with each other at 50°C with moderately stirring to provide a solution 1 containing a reactive polymer having a photopolymerizable functional group.

[0061]

20 The resultant reactive polymer has Tg of 0°C, and 5% by mole of methacryloyl group on its side.

[0062]

Formulation II

solution 1 of reactive polymer	100 parts by weight
tricyclodecane diacrylate	30 parts by weight
25 1-hydroxycyclohexyl phenyl ketone	1 part by weight

The above Formulation II was homogeneously dissolved to give a

mixture, which was applied onto a release sheet (thickness: 75 $\mu\text{m}$ ; trade name: No. 23, available from Fujimori Kogyo) and dried to form a photo-curable adhesive sheet (layer) of thickness of 25 $\pm$ 2 $\mu\text{m}$ . Thus, the resultant photo-curable adhesive sheet had the total thickness of 100 $\pm$ 2 $\mu\text{m}$ .

5 [0063]

<Preparation of one optical information recording substrate having reflective layer>

A photo-curable transfer sheet was prepared in the same manner as in the photo-curable adhesive sheet. The resultant photo-curable transfer 10 sheet having release sheet had thickness of 100 $\mu\text{m}$ , which is thicker than the adhesive sheet.

[0064]

The photo-curable transfer sheet was depressed on an unevenness surface of a stamper having the uneven surface as pits using a roller made 15 of silicone rubber under load of 2kg to form a laminate in which the shape of the uneven surface was transferred to a surface of the photo-curable transfer sheet.

[0065]

Subsequently, the photo-curable transfer sheet of the laminate was 20 exposed to UV-rays of a metal-halide lamp under the condition of an integrated amount of light of 1,000mJ/cm<sup>2</sup> and as a result, the transferred layer (photo-curable sheet) was cured.

[0066]

The stamper was peeled from the laminate. Silver alloy was spattered 25 on the uneven surface of the cured photo-curable layer (optical information recording substrate) to form a semitransparent reflective layer of

silver alloy. Thus, an optical information recording substrate having reflective layer was prepared.

[0067]

<Preparation of the other optical information recording substrate having re-

5      flective layer>

Melt carbonate was poured into a mold having an uneven surface as pits and solidified to form an optical information recording substrate having thickness of 1,100 $\mu\text{m}$ . Aluminum was spattered on the uneven surface of the optical information recording substrate to form a reflective layer 10 of Al. Thus, the other optical information recording substrate having reflective layer was prepared.

[0068]

<Preparation of optical information recording medium>

The two optical information recording substrates prepared above 15 were bonded to each other through the photo-curable adhesive sheet prepared above such that the two reflective layers faced each other to give a laminate, and the laminate was exposed to UV-rays of a metal-halide lamp under the condition of an integrated amount of light of 1,000mJ/cm<sup>2</sup> and as a result, the photo-curable adhesive sheet was cured. Thus an optical in- 20 formation recording medium (DVD) was prepared.

[0069]

[Example 2]

<Preparation of photo-curable adhesive sheet>

(Preparation of reactive polymer)

25      Formulation I'

n-hexyl methacrylate

50 parts by weight

2-hydroxyethyl methacrylate	50 parts by weight
benzophenone	5 parts by weight
toluene	30 parts by weight
ethyl acetate	30 parts by weight

5        A mixture of the above Formulation I' was heated to 60°C with moderately stirring to initiate the polymerization, and stirred at this temperature for 10 hours to provide acrylic resin having a hydroxyl group on its side chain. Then, 50 parts by weight of Calens MOI (2-isocyanatoethyl methacrylate; available from Showa Denko K.K.) was  
10      added to the solution of the acrylic resin, and reacted with each other at 50°C with moderately stirring to provide a solution 2 containing a reactive polymer having a photopolymerizable functional group.

[0070]

15      The resultant reactive polymer has Tg of 5°C and 50% by mole of methacryloyl group on its side.

[0071]

#### Formulation II'

solution 2 of reactive polymer	100 parts by weight
1,6-hexanediol dimethacrylate	10 parts by weight
20      1-hydroxycyclohexyl phenyl ketone	1 part by weight

25      The above Formulation II' was homogeneously dissolved to give a mixture, which was applied onto a release sheet (thickness: 75µm; trade name: No. 23, available from Fujimori Kogyo) and dried to form a photo-curable adhesive sheet of thickness of 25±2µm. Thus, the resultant photo-curable adhesive sheet had the total thickness of 100±2µm.

[0072]

One and the other optical information recording substrates and optical information recording medium were prepared in the same manner as described in Example 1 except for using the above photo-curable adhesive sheet. Thus DVD was obtained.

5 [0073]

[Comparison Example 1]

An optical information recording medium was prepared in the same manner as described in Example 1 except for performing the preparation of one optical information recording substrate and the recording medium in  
10 the following manner:

[0074]

<Preparation of one optical information recording substrate having reflective layer>

Melt carbonate was poured into a mold having an uneven surface as  
15 pits and solidified to form an optical information recording substrate having thickness of  $100\pm2\mu\text{m}$ . Silver alloy was spattered on the uneven surface of the optical information recording substrate to form a semitransparent reflective layer of silver alloy. Thus, one optical information recording substrate having reflective layer was prepared.

20 [0075]

<Preparation of optical information recording medium>

A liquid curable adhesive (SD-661; available from DAINIPPON INK AND CHEMICALS, INC.) which is commercially available was applied onto one of the two optical information recording substrates prepared  
25 above. The two optical information recording substrates were bonded to each other through the adhesive such that the two reflective layers faced

each other to give a laminate, and the laminate was exposed to UV-rays whereby the adhesive sheet was cured. Thus an optical information recording medium (DVD) was prepared.

[0076]

5 <Evaluation of optical information recording substrate and medium>

(1) Light transmittance (wavelength of 380 to 800 nm)

Light transmittance of the resultant photo-curable adhesive sheet is measured in the wavelength of 380 to 800 nm according to JIS K6717.

Light transmittance of 70% or more is marked as ○, and Light transmitt-

10 tance of less than 70% is marked as ×.

[0077]

(2) Light transmittance (wavelength of 380 to 420 nm)

Light transmittance of the resultant photo-curable adhesive sheet is measured in the wavelength of 380 to 420 nm according to JIS K6717.

15 Light transmittance of 80% or more is marked as ○, and Light transmittance of less than 80% is marked as ×.

[0078]

(3) Roughness of land portion

A land portion of an uneven surface on which pits were formed is evaluated on its smoothness using AFM (atomic force microscope). Land portion having sufficient smoothness is marked as ○, and land portion having poor smoothness is marked as ×.

[0079]

(4) Readout of signals

25 The information of the resultant optical information recording medium is read out using a laser beam of wavelength of 405nm to obtain its

wavy pattern. This wavy pattern is compared with that of the stamper. The wavy pattern of the medium coincident with that of the stamper is marked as ○, and the wavy pattern of the medium little coincident with that of the stamper is marked as ×.

5 [0080]

The obtained results are shown in Table 1.

[0081]

Table 1

	Example 1	Example 2	Com. Example 1
Light transmittance (380-800nm)	○	○	○
Light transmittance (380-420nm)	○	○	○
Roughness of land	○	○	×
Readout of signals	○	○	×

10 [Effect of the invention]

[0082]

As shown above, the photo-curable adhesive sheet according of the invention is depressed on an uneven shape of an optical information recording substrate to precisely follow the uneven surface and adhere to it.

15 Thus the resultant optical information recording substrate has a signal surface (uneven surface) to which the adhesive layer (sheet) precisely adheres, and is free from adverse effect on the signal surface by the adhesion. Accordingly, the resultant optical information recording medium scarcely brings about occurrence of errors when the information (signals) is read  
20 out.

## [0083]

Further, in the adhesion procedure in the preparation of the optical information recording substrate of the invention, the photo-curable adhesive sheet used is softened and deformed and allowed to closely adhere to the uneven surface, and then cured. Therefore, optical information recording substrates can be bonded to each other by even a thin layer having a thickness of 300 $\mu\text{m}$  or less. Further, the photo-curable adhesive sheet of the invention has high transparency compared with conventional UV curable resin, and furthermore has excellent dimensional stability due to less cure shrinkage than a conventional UV-curable resin. Hence, the resultant optical information recording medium prepared using the adhesive sheet does not suffer from deformation such as warpage.

## [0084]

For example, the photo-curable adhesive sheet of the invention has a small thickness (e.g., 25 $\pm$ 2 $\mu\text{m}$ , nonuniformity of  $\pm$ 2 $\mu\text{m}$ ) as an adhesive layer, and therefore the adhesive sheet shows excellent precision in the thickness compared with that of an adhesive layer formed by spin-coat-application of a conventional UV curable resin liquid, the application bringing about thickness-nonuniformity of  $\pm$ 5 $\mu\text{m}$ . Such less thickness-nonuniformity can bring about enhancement of the dimensional stability.

## [0085]

Since the photo-curable adhesive sheet of the invention has the above excellent characteristics, it is apparent that the sheet is useful in not only the preparation of the resultant optical information recording medium but also the bonding operation in various fields.

[Brief Description of the Drawings]

[Fig. 1] Fig. 1 is a section view showing an example of an embodiment of a photo-curable adhesive sheet according to the present invention.

5 [Fig. 2] Fig. 2 is a section view showing an example of a process for the preparation of the optical information recording medium according to the invention.

10 [Fig. 3] Fig. 3 is a section view showing another example of a process for the preparation of the optical information recording medium according to the invention.

[Fig. 4] Fig. 4 is a schematic view for explaining a depressing method using a device according to a double vacuum chamber system.

[Brief Description of the reference numbers]

- 15 11: Photo-curable adhesive sheet  
12a, 12b: Release sheet  
23: Reflective layer  
21, 24: Optical information recording substrate  
25: Semitransparent reflective layer

[Title of Document] Abstract

[Abstract]

[Object]

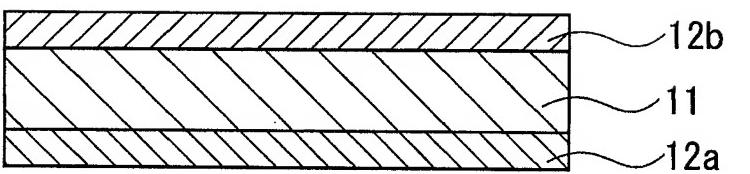
The object of the invention is to provide a photo-curable adhesive  
5 sheet used is easily and precisely allowed to closely adhere to uneven surface of a stamper for preparing a substrate of an optical information recording medium by pressing to the uneven surface, and a process for preparing optical information recording medium by using the sheet.

[Means for Achieving the Object]

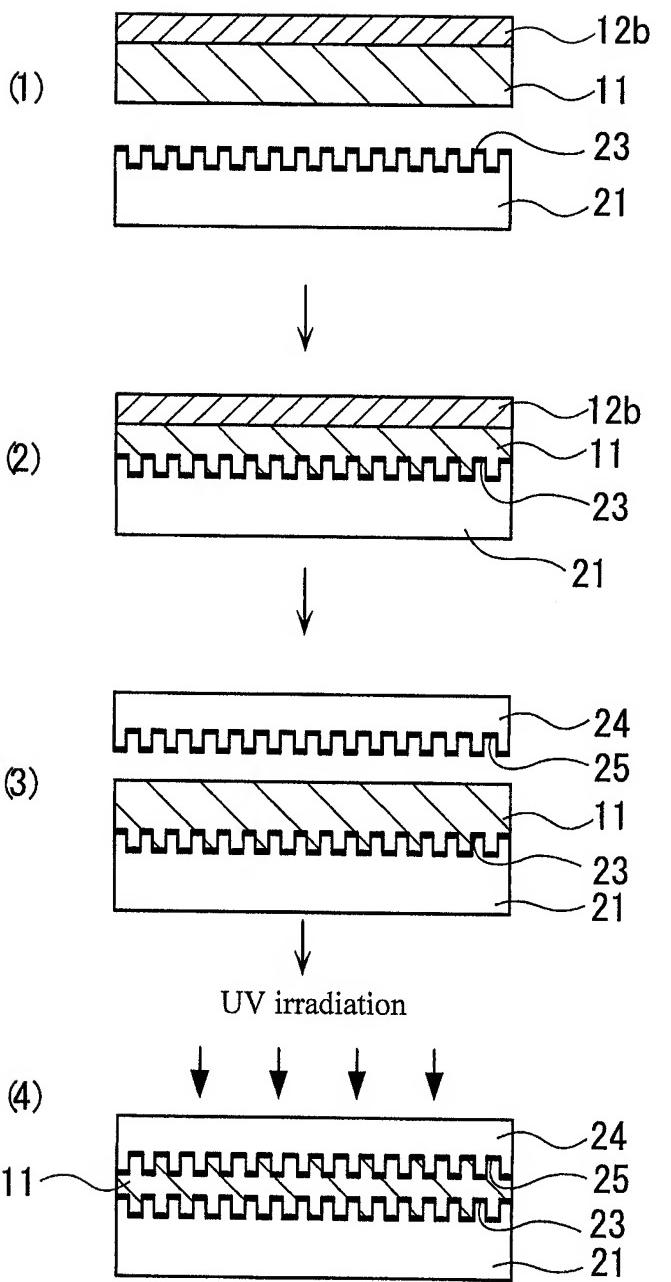
10 A photo-curable adhesive sheet comprising a photo-curable composition which comprises a reactive polymer having a photopolymerizable functional group and which has a glass transition temperature of not more than 20°C, the photo-curable adhesive sheet having a light transmittance of not less than 70% in a wavelength range of 380 to 420 nm; and an optical  
15 information recording substrate and medium obtained by using the sheet.

[Selected Figure] Fig. 2

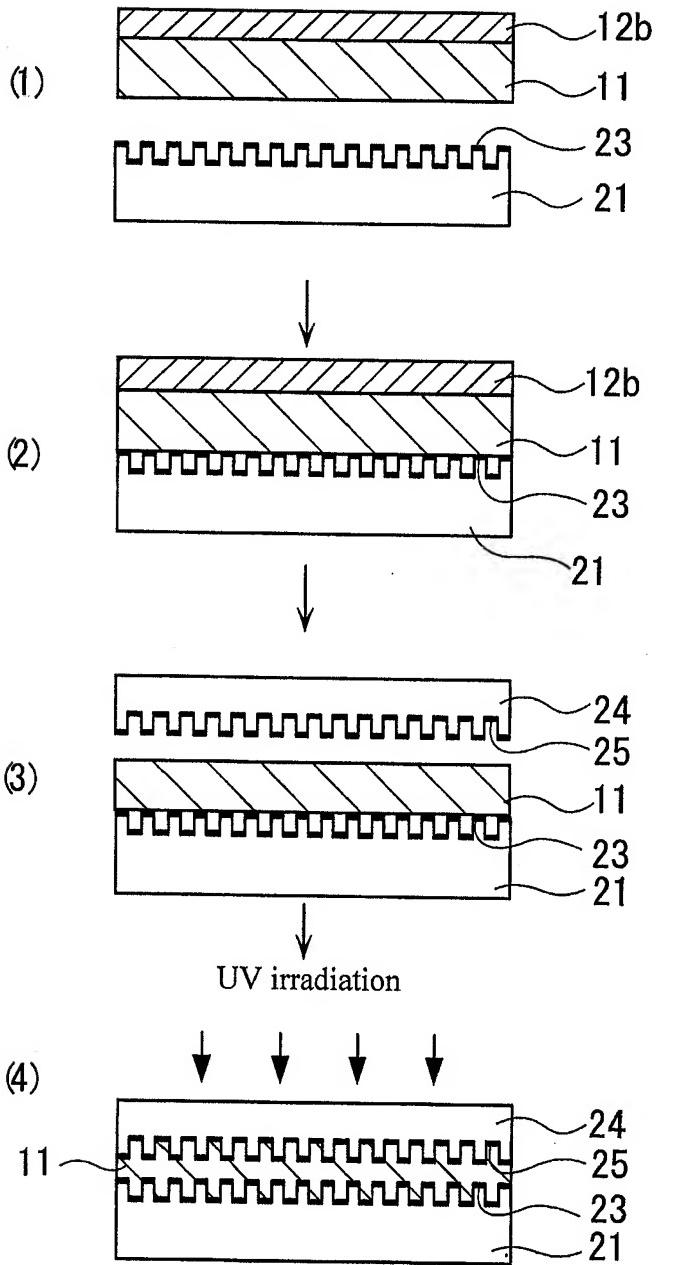
【Fig. 1】



【Fig. 2】



【Fig. 3】



【Fig. 4】

